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PROGRESS IN PHOTOCHEMICAL AIR QUALITY SIMULATION MODELING

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INTRODUCTION

Over the past 15 years we have witnessed the development of atmospheric dispersion models which include increasingly complex approximations of photochemistry. Unlike existing Gaussian or analytic schemes, the models simulate atmospheric chemical reactions, some of which act at very rapid rates, and therefore involve time steps much shorter than are generally regarded as necessary for transport and dispersion of inert species. The computational demands rise rapidly with the number of chemical species considered, and it is not unusual to see present-day models for urban and regional scales having computer simulation speeds comparable to the real-world events. Computer restrictions have tended to engender decisions to diminish the spatial resolution of the models in order to increase the number of species and reactions treated.

We have recognized for a number of years that within the class of existing or conceived photochemical air quality simulation models (PAQSM's) there are only several basic approaches. The United States Environmental Protection Agency (U.S. EPA) in the mid-1970's reviewed the various urban scale models which were in existence and chose three, embodying distinct approaches, for further refinement, development, and evaluation. In the case of the EPA, the ultimate goal was to provide regulatory tools for control of photochemical pollutants, O_3 in particular. The models

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16. ABSTRACT

Within the class of existing or conceived photochemical air quality simulation models (PAQSM's) there are only several basic approaches. The United States Environmental Protection Agency (U.S. EPA) in the mid-1970's reviewed the various urban scale models which were in existence and chose three, embodying distinct approaches, for further refinement, development and evaluation. In the case of the EPA, the ultimate goal was to provide regulatory tools for control of photochemical pollutants, O_3 in particular. Shortly thereafter, work also began on a regional-scale (1000 km) photochemcial model to be used initially in the northeastern U.S. Interest in larger scales derived from a growing awareness that elevated O_3 levels may be found over large regions and at considerable distances from urban centers.

The purpose of this paper is to describe the PAQSM's emerging from research and development projects of the U.S. EPA. The presentation will not attempt a detailed discussion of chemical mechanisms or numerical schemes. Rather, emphasis will be placed on the models' basic structures, data requirements, computer requirements, and problems encountered in applying them. Also, two major field programs will be described which have been conducted by EPA to support testing and evaluation of the models.

17. KEY WORDS AND DOCUMENT ANALYSIS

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were to be used to predict the problems created by new sources or the benefits of controlling existing sources. Shortly thereafter, work also began on a regional-scale (1000 km) photochemical model to be used initially in the northeastern U.S. Interest in larger scales derived from a growing awareness that elevated O_3 levels may be found over large regions and at considerable distances from urban centers (Wolff et al., 1977). The goal of the regional modeling effort was again to allow defensible regulatory decisions, but the complexity of the problem and its immense resource requirements precluded study of several alternate approaches as was done with the urban models.

The purpose of this paper is to describe the PAQSM's emerging from research and development projects of the U.S. EPA. The presentation will not attempt a detailed discussion of chemical mechanisms or numerical schemes. Rather, emphasis will be placed on the models' basic structures, data requirements, computer requirements, and problems encountered in applying them. Also, we will describe the two major field programs which have been conducted by EPA to support testing and evaluation of the models.

COMPONENTS OF MODEL SIMULATIONS

PAQSM's may differ in complexity and therefore data and resource requirements through deliberate choices of the builders. For example, restriction to a simple box-model makes spatial resolution of emissions and winds unnecessary. Furthermore, data requirements will vary greatly depending on the goal of the simulations. If model development and evaluation are planned, then the data needs are extensive. On the other hand, if the model is already viewed as a reliable operational tool, data needs are greatly reduced as climatological scenarios may be used for worst-case and average events. In the latter case, predicted pollutant levels are only checked for reasonableness against previous records and not against specific observations. Photochemical air quality simulations share similar attributes, and requirements, regardless of the particular model used. Generally, all PAQSM's are logical frameworks which synthesize information from meteorology, sources, and air quality and predict the photochemical pollutant concentrations consistent with expressions for governing physical laws. The simulation is therefore an interplay of four equally important components.

PHYSICAL LAWS: Physical laws are expressed as mathematical equations governing atmospheric motions (transport and diffusion) and the chemical kinetics. The equations enforce mass conservation and promote appropriate chemical transformations in a manner consistent with theoretical considerations and observational data.

Kinetic mechanisms generally have been developed by comparing numerical predictions to concentrations resulting from carefully controlled smog chamber experiments. It is important to note that equations of both the meteorological and chemical aspects of the problem are approximations. For example, mixing rates are usually difficult to prescribe, and hydrocarbon (HC) species must be lumped in terms of reactivity classes.

SOURCE EMISSIONS: Pollutant emissions are an important component in the application of a PAQSM and must be compiled on spatial and temporal scales comparable to the resolution inherent in the model structure. Photochemistry involves a strong diurnal cycle which dictates a temporal resolution on the order of 1-h in the emissions inventory. Inventories usually are divided into the source types of area, point, and line. Area sources include many diffuse, population-related sources such as those associated with space heating or residential automotive traffic. Point sources are large, identifiable sources such as power plants, and line sources refer to major automotive thoroughfares. A typical inventory for a large city may include several hundred point sources. Unless the model has the unusual capacity to treat line sources, they may be incorporated into the area source inventory. For PAQSM's the emphasis is usually on developing emissions for HC, NO_x, and CO, although mechanisms generally include other species such as SO_2 and SO_4. The HC emission is broken into reactivity classes consistent with the kinetic mechanism (e.g., olefins, paraffins, aromatics, formaldehyde, and other aldehydes). Also, the NO_x must be appropriately split between NO and NO_2.

METEOROLOGICAL FACTORS: Meteorological factors account for the transport and diffusion of pollutants and influence reaction rates in the chemical mechanism. These data are introduced into the model from observations and are not predicted. Important parameters include the wind field, inversion height, atmospheric stability, solar radiation, water vapor, and temperature.

AIR QUALITY MEASUREMENTS: Ambient concentration measurements are needed to set initial conditions, boundary conditions, and as evaluation data. Photochemical simulations usually begin with an initial set of observations used to assign concentrations of important O_3 precursors, HC and NO_x, within the modeled domain. As an alternate strategy, the model could generate its own initial state by allowing sufficient time for emissions to establish suitable concentration levels. However, this strategy is usually impracticable when considering other aspects of the problem such as ventilation rates, especially on the urban-scale. Boundary conditions refer to the concentrations assigned at the extremities of the modeled region, both in the horizontal and vertical. Concentrations at the upwind boundary of a grid-model domain are of obvious impor-

tance. Moreover, the concentrations aloft may also play a role as they are entrained during the diurnal growth of the mixed layer. Ambient data must have a spatial and temporal resolution at least comparable to the model output if evaluation of the model is to be attempted.

URBAN MODELS

The U.S. EPA currently has interest in three urban models embodying distinctly different approaches and levels of complexity. Those models are:

Photochemical Box Model (PBM) - a single cell Eulerian model constructed by EPA.

Lagrangian Photochemical Model (LPM) - a multi-level parcel model originally developed by Environmental Research and Technology, Inc.

Urban Airshed Model (UAM) - a multi-level, Eulerian grid model originally developed by Systems Applications, Inc.

The versions of the models at EPA have been structured to easily use urban data compiled during the Regional Air Pollution Study (RAPS) and have been subject to continuing modifications. The RAPS will be discussed in a later section. In this section, the basic frameworks and requirements of the models will be surveyed.

Photochemical Box Model

The Photochemical Box Model (PBM) is a single cell Eulerian air quality model whose purpose is to simulate the transport and chemical transformation of air pollutants in smog prone urban atmospheres. The model's domain is set in a variable-volume, well-mixed reacting cell where the physical and chemical processes responsible for the generation of O_3 by its HC and NO_x precursors are mathematically created. These processes include the transport and dispersion of pollutant species through the cell, the injection of primary precursor species by emission sources, and the chemical transformation of the reactive species into intermediate and secondary products. They are schematically illustrated in Figure 1. In a typical application of the model, the horizontal length scale of the single cell is about 20 km and the vertical scale is time-varying, equivalent to the depth of the mixed layer. The model domain is centered on the city such that the area encompasses most of the major emissions sources. Source emissions are assumed to be distributed uniformly across the surface face of the cell.

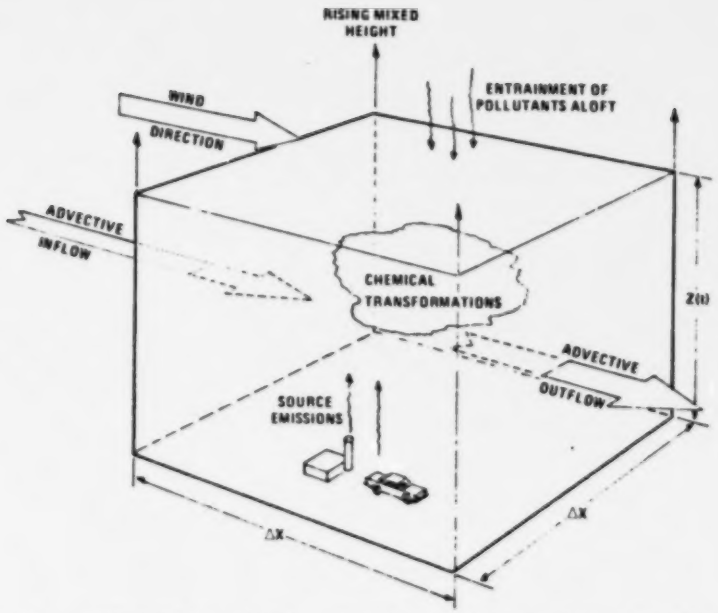

Fig. 1. Schematic of the domain and processes in the Photochemical Box Model.

While the data requirements of the PBM are not particularly rigorous, three data preprocessors must be executed prior to each model simulation. The first accesses the 1-h average air quality and meteorological data base forming the initial concentrations of participating species, determining the 1-h average wind vectors that guide transport through the model domain, and choosing the hourly updated concentration of species at the upwind boundary of the box. The first preprocessor also forms similar spatial averages of concentrations of monitored pollutant species for comparison with model predictions.

A function of the second preprocessor is to determine 10 min average values of total solar radiation from pyranometer measurements. From these values the time-varying photolytic rate constants can be determined through a series of empirical and theoretical relationships. The temporal resolution of these data is greater than for most other data because of the rapid chemical reactions which are associated with these rate constants. This preprocessor also calculates the diurnal growth pattern of the mixed layer, or the depth of the model domain. To perform this calculation, the morning minimum and afternoon maximum mixed layer heights are provided by the user. These heights were determined in the RAPS application by studying the vertical structure of the temperature and moisture from radiosondes. Releases occurred

throughout the day at 6-h intervals beginning 1 h before sunrise. The model fits a piece-wise linear function between the specified minimum and maximum depths that simulates the actual mixed layer growth as observed by acoustic sounder, lidar, and radiosonde techniques. Ten-minute averages of mixed layer height are formed for the PBM; photolytic rate constants for the corresponding time period are integrated through this depth.

The third preprocessor is responsible for forming hourly emissions source terms for CO, NO, NO_2, and non-methane HC. Even though the emissions may have spatial resolution, all source emissions, both area and point, within the model domain are summed to provide a total emission rate for each primary pollutant species at every hour of simulation.

The PBM may be executed when the data files from the three preprocessors have been created. The only other relevant information needed at this time is the concentration of O_3 at the top boundary of the modeling domain, that pollutant having been transported into the area by winds aloft during the night. This concentration is determined from the O_3 measurements at the far upwind surface monitoring sites after the nocturnal temperature inversion has eroded and the air aloft mixes down through the atmosphere to the surface. Concentrations of other pollutants at the top of the modeling region are assumed to be negligible. The simulation is typically started at 0500 CST and continues through 1700 CST.

The PBM contains a chemical kinetic mechanism with 36 reactions and 27 reactive species (Demerjian and Schere, 1979). The set of equations describing the rates of change of the concentrations of these species is numerically solved at time steps on the order of 10 min. From these solutions the model determines the 1-h average predicted concentration of each modeled species.

Lagrangian Photochemical Model

The Lagrangian Photochemical Model (LPM) was developed by Environmental Research and Technology, Inc. of Westlake Village, California and adapted under contract with EPA for use with the RAPS data base. (The LPM is essentially identical to the general-use model named ELSTAR.) The LPM envisions a portion of the atmosphere as an identifiable parcel which can be tracked from early morning to the late afternoon. As the parcel moves over the various emissions sources, pollutants are assimilated, vertically mixed, and subjected to photochemical reactions in the presence of solar radiation (see Figure 2). The LPM is attractive relative to grid models in that it is fairly simple to execute and uses a moderate amount of computer time. On the other hand, the LPM

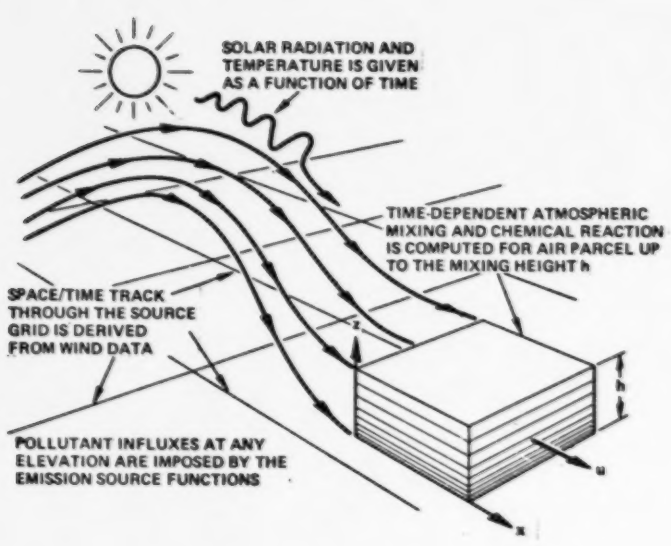

Fig. 2. Schematic of the modeling concepts of the Lagrangian Photochemical Model.

calculates concentrations only within a parcel and not over a complete spatial field.

The LPM is executed using a series of program modules. They are METMOD, EMMOD, and KEMOD, sequentially performing calculations on meteorology and air quality, emissions, and photochemistry. The input and running procedures described by Lurmann et al. (1979) have been generally followed, although some modifications were deemed necessary as more experience with the model was acquired (Lurmann, 1980; 1981).

The first step in setting up a simulation is to determine the starting point of a parcel so that it will arrive at a specified point at an assigned time. A backward trajectory can be generated by the METMOD. The parcel trajectory is usually determined by $1/R^2$ weighting of winds from the closest three stations. However, experience with wind data suggests that even closely situated stations can show large, unexplained differences in wind vectors from time to time. Reliance on a single anomalous station, if the parcel has a close approach, can create an erratic trajectory. To eliminate the possibility of such vagaries, it may be prudent to compute a single resultant wind vector from the wind station network and assign it to all stations for a given hour. The resultant gives a general movement of the air mass over the region.

Because of the difficulty in solving the chemistry set with minimal solar radiation, the start time of a parcel (which is on the hour) must be at least 10 min past local sunrise. Once the start position is set, the METMOD is run in a forward-trajectory mode until 1800 CST or the parcel leaves the region.

Mixing heights are computed from radiosondes released approximately 1 h before sunrise and at 6-h intervals thereafter. The temperature and wind profiles from the balloons are processed automatically by the model to give vertical eddy diffusivities throughout the day. The trajectory information from METMOD is stored and is available to EMMOD to enable that module to obtain the emissions that the parcel will encounter. METMOD also stores the observed pollutant concentrations along the trajectory using a $1/R^2$ weighting scheme on the closest three stations. These observations are available for setting initial conditions in the parcel and later comparisons with the model predictions. In most cases, the parcel starts in a relatively clean rural environment, and levels of HC, NO, and NO_2 are assumed to decrease with height according to an assigned formula. On the other hand, ozone is depleted near the ground at night; thus, the initial O_3 increases from the surface to a value at 400 m which is equal to the observed 1000-1200 CST surface concentration upwind of the city (Shreffler and Evans, 1982).

The EMMOD creates a record of the emissions entering the parcel as it traverses its trajectory. These emissions are used by the KEMOD to simulate the photochemical reactions which will take place as a function of time, yielding concentrations of 39 chemical species at 30-min intervals. A total of 65 reactions are modeled.

Urban Airshed Model

The Urban Airshed Model (UAM) is a three-dimensional (3-D) grid-type, or Eulerian, PAQSM developed by Systems Applications, Inc. (SAI) of San Rafael, California. The structure of the model consists of a latticework array of cells (see Figure 3), the total volume of which represents an urban-scale domain and in which the physical and chemical processes responsible for photochemical smog are mathematically simulated. These processes include the advection of pollutant species through the modeling domain, the species entrainment from aloft by a growing mixed layer, the diffusion of material from cell-to-cell, the injection of primary source emissions into the modeled volume, and the chemical transformations of reactive species into intermediate and secondary products. The horizontal dimensions of each cell are constant but the heights of the cells vary throughout a model simulation as the depth of the mixed layer in the UAM changes accordingly.

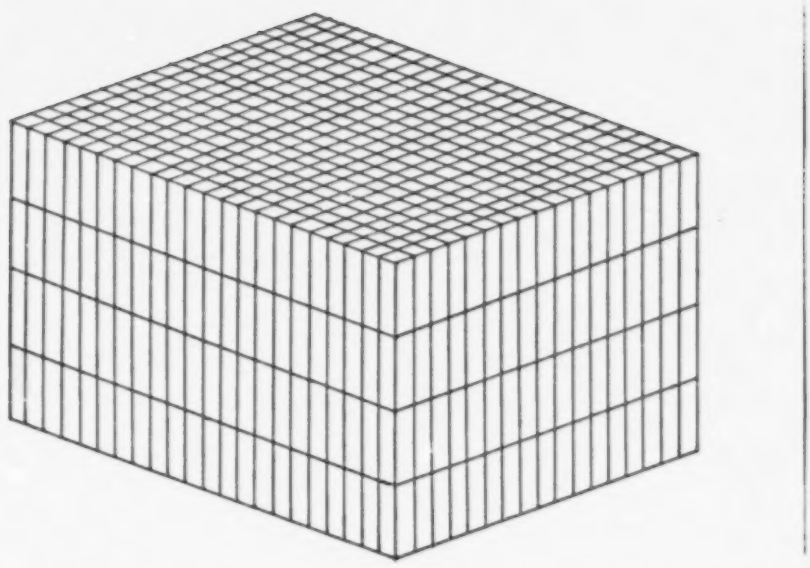

Fig. 3. Schematic of the grid domain used in the Urban Airshed Model.

In typical applications, the area modeled is about 60 x 60 km, and each individual cell is 4 km on a side in the horizontal. Vertically, there are four layers of cells in total; the bottom two layers simulate the mixed layer and the top two represent the region immediately above the mixed layer. The 3-D grid model is a sophisticated type of PAQSM and provides both spatially and temporally resolved concentration predictions. Thus, the UAM attempts to estimate the 1-h average observed concentration of a pollutant species at each monitoring site within the model domain.

The package of computer programs constituting the UAM actually contains 12 data preprocessing programs as well as the simulation model. The data requirements for applying the model are rather intensive. The preprocessors (PP's) access surface-based, hourly air quality and meteorological data base, the upper air pibal and radiosonde data, and the source emissions inventory for the necessary parameters, and process the parameters as required by the simulation model. A brief description of the PP's follows in order to convey a sense of the UAM data requirements.

The chemistry PP sets up the rate constants and other parameters related to the chemical kinetic mechanism within the model. This mechanism, named Carbon Bond II, is a state-of-the-art set of chemical reactions describing the NO_x-HC-O_3 species interactions

within a photochemically active atmosphere. The mechanism was developed at SAI by Whitten et al. (1980). The terrain PP describes the spatially varying types of surfaces within the modeling domain, reflecting the transition from urban to suburban and finally to rural land uses. The spatial resolution of these land forms should be on the same scale as the grid cell size. Both the chemistry and terrain PP's need only be executed once for a particular model application. All of the remaining PP's, however, must be executed prior to each model simulation within the application.

The diffusion-break and region-top PP's describe the variations in the mixing heights and modeling region depths, respectively, over the domain during the course of the simulation. Hourly values of the mixing height must be supplied at representative locations on the grid. It is the responsibility of the user to specify these depths in a meaningful manner from upper air soundings or equivalent data. The meteorological PP describes the temporal variation of vertical temperature gradient, stability class, atmospheric pressure, water vapor concentration, and the NO_2 photolysis rate constant. Values for these parameters do not vary spatially in the model. The top-concentration PP specifies the concentrations of principal species at the top of the modeled region throughout the simulation. In most cases these concentrations will be close to the clean air background values for these species, although substantial concentrations of O_3 can result from advection over the region. Its value is determined from the O_3 measurements at the far upwind surface monitoring sites after the nocturnal temperature inversion has eroded and the air aloft mixes to the surface.

The air quality, temperature and wind speed PP's all require data from a surface monitoring network. Hourly averages of observed species concentrations are objectively interpolated across the model grid by the air quality PP to produce a field of initial concentrations for the UAM. Typically this initial field is applied near sunrise. As the density of monitoring locations increases, the reliability of the objective interpolation does likewise. The temperature PP produces gridded fields of surface temperature at each hour of model simulation. These are required for both the wind and point source PP's. Finally, the wind PP assimilates all available surface and upper air wind speed and direction measurements and produces a gridded field of u and v wind components at each of the model's four vertical levels. This objective wind field analysis routine was developed by Anderson of SAI (Killus et al., 1977). Vertical wind velocities are calculated internally by the UAM from mass continuity considerations. Anderson's interpolation routine smooths the data in such a way so as to eliminate unrealistic vertical motions. It simulates the mesoscale urban circulation patterns through the use of the surface temperature

patterns. The boundary concentration PP determines the modeled species concentrations along each of the domain's borders for each hour of model simulation. A vertical concentration gradient is also described.

The last two PP's needed before the model is executed deal with source emissions. The area sources, including highway and line sources, and the point sources are treated separately. Hourly emission rates of all primary pollutants for each grid are calculated. Organic HC emissions must be distributed into particular structural classes.

Each PP generates a data file required by the UAM. Model simulations begin at 0500 CST and continue through 1700 CST. The model numerically calculates the rates of change of species concentrations at time steps on the order of several minutes, and from these determines the 1-h average predicted concentrations.

Status of the Urban Models

The three models discussed in this section are viewed by EPA as being in final form. That is, substantial breakthroughs in chemical mechanisms or the compilation of a clearly superior data base will be needed before revisions would be considered. Although the model preprocessors are geared to easy use of the RAPS data, it would not be difficult to adapt the models to other locations and other forms of data. In the next section the RAPS data set will be described along with some results of model applications. Before proceeding, we review some problems which arose during the testing of the models and look at estimated resource requirements.

Because of its relative simplicity, the PBM is attractive for use where resources are limited and results do not need to be highly detailed. With this model, it should be noted that emissions control strategies are difficult to target on particular sources because of the lack of spatial resolution. Also, the highest O_3 concentrations in the domain, upon which decisions may be more critically centered, are lost in the averaging assumptions. Finally, the simplified wind field (a single vector for each hour) and large size to the Eulerian domain make the model best suited for low-wind or stagnation episodes. These, of course, are usually conditions most favorable to high O_3 concentrations.

Compared to the UAM, the LPM may seem less resource intensive. However, the data requirements are nearly identical. The LPM may be used along a single trajectory, perhaps that trajectory encountering the observed maximum concentration, and therefore use much less computer time. If multiple trajectories are executed to display

concentrations over the region, such as the UAM does in a single simulation, then computer resources and preparation time will grow accordingly.

A number of problems have become apparent in the development and testing of the models. For example, the LPM seems quite sensitive to initial conditions since the parcel retains all pollutants. Thus, care must be taken to put realistic vertical distributions of precursors in the parcel at the beginning of the simulation day. The LPM has five vertical levels in which to distribute the pollutants. As originally conceived the model did not include any lateral diffusion, and this produced unrealistically high O_3 concentrations since all emissions were retained in a fixed volume. The model now allows the parcel to expand laterally in a manner commensurate with empirically determined diffusion. Whereas this feature considers dilution of a parcel when beyond the high emissions area, it introduces a problem in setting side boundary condition concentrations representative of the air that is entrained into the parcel. Our approach has been to set the initial parcel size about equal to the downtown area of highest emissions (5 x 5 km for RAPS) and treat the entrained air as having background values of HC and NO_x. The parcel is viewed as a segment of the urban plume. However, large power plant sources are liable to present conditions seriously in conflict with that assumption from time to time. This example evinces a fundamental truth about all of the PAQSM's; there is an element of art in their application which is unlikely to be eliminated without jeopardizing the models' flexibility in treating different situations.

The UAM as originally constructed suffered from substantial numerical diffusion. We believe this potential problem should be thoroughly investigated in any grid model of this sort. The tendency of the UAM to underestimate ozone maxima in our tests (see results in next section) was suspected of being due to the spurious diffusion. However, the underestimation was only slightly improved by a new and accurate numerical scheme, which, incidentally, increased computer time by 15 percent (Schere, 1982).

The choice of which particular model to use in a specific application involves not only the accuracy of the model but also the resources required to operate it. The models discussed here have resource requirements correlated with their level of complexity. In terms of man-months needed to set up a single day simulation and computer time expended (minutes of CPU on a Univac 1100/82) the approximate requirements are:

PBM	0.15 man-month	1 minute CPU
LPM	0.20 man-month	10 minutes CPU
UAM	0.50 man-month	110 minutes CPU

The initial man-month investment needed to become adequately familiar with the model perhaps exceeds the simulation set-up time by at least a factor of 10.

The three models discussed here have all shown themselves to be acceptable tools for analysis of urban O_3 air quality. The specific configuration of an application along with the quantity and quality of related data and resources available to the user must all be considered in the final selection of a model. For an indication of average O_3 air quality in an urban area under stagnation conditions or as a screening method for a more complex model, the PBM is appropriate. The choice of a trajectory model, such as the LPM, or a grid model, like the UAM, might well be decided by resource requirements or by the number of proposed simulations. In any event, the user of any of these models must have a strong scientific background and exercise extreme care in implementing the air quality simulations.

THE RAPS AND URBAN MODEL EVALUATION

Beside the physical laws expressed in the PAQSM, essential components of air quality simulations include emissions, meteorological and ambient concentration data. In this section we describe an extensive field effort for gathering data on an urban scale and some of the modeling results based on that data.

The Regional Air Pollution Study (RAPS) was carried out in the St. Louis area during 1974-1977. St. Louis is a city of negligible topographic relief in the central U.S.; about 2.3 million people reside in the metropolitan area. Although augmented by a variety of special studies, the principal data base was gathered by a 25 station network comprising the Regional Air Monitoring System (RAMS). Measurements included both meteorological parameters and pollutant concentrations. The purpose of RAPS was to develop a data base for testing numerical air quality simulation models and especially models involving the complex photochemical reactions leading to O_3 formation. The RAPS expenditures totaled about $25 million over a five-year period, and the data base remains the most comprehensive available.

RAMS measurements, recorded as 1-min averages, included wind speed and direction, temperature, vertical temperature difference, solar radiation and concentrations of O_3, CO, HC, NO_x and SO_2. The sampling manifold used by all gaseous monitors had an intake at about 4 m from the ground. Figure 4 shows the layout of the RAMS network. A more detailed description of the RAPS program and the RAMS instrumentation is given by Schiermeier (1978).

Fig. 4. The St. Louis area with locations of the RAPS stations 101-125.

The 1-min average values of all parameters were objectively screened. The screening excluded values which were null, part of a calibration set, taken during a period of excessive drift, outside instrument limits, taken under abnormal instrument status, or indicating persistence. The remaining data were used to form 1-h averages, and the 1-h average archive was generally the one used in model evaluation.

Overviews of the RAPS O_3 and NO_x data are given by Shreffler and Evans (1982) and Shreffler (1982). Figure 5 displays a 4-mo record of ozone taken in mid-1975. The time series are for the inflow concentration (that from upwind rural areas) and the maximum 1-h concentration recorded at any station in the network. The locations of the maxima, especially the strong peaks, usually show

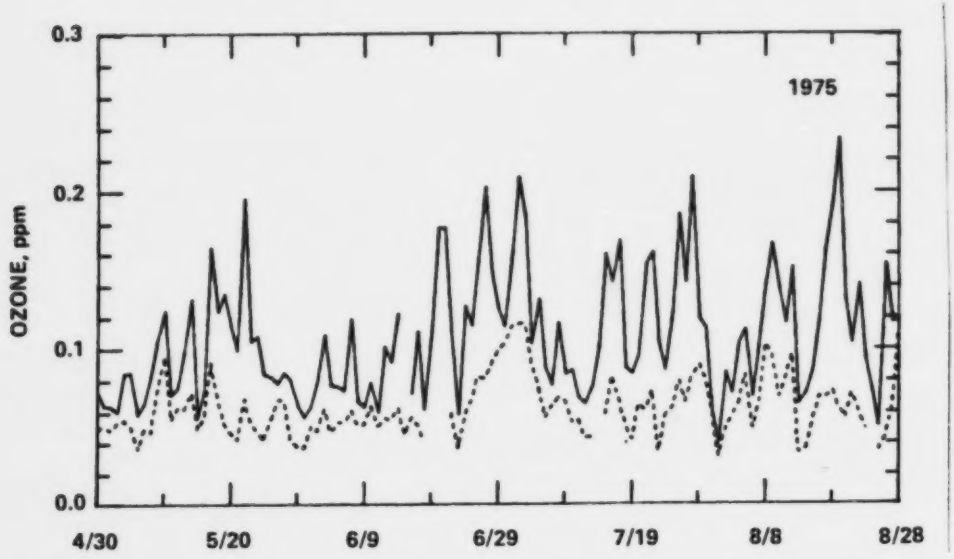

Fig. 5. Time series of the daily maximum 1-hr ozone concentration (solid) and the daily rural inflow ozone concentration (dashed) for May - August 1975 as measured in the RAPS.

clear downwind relations to high emissions areas. The differences between concentrations in the two series may be ascribed to net enhancement of O_3 levels due to the urban emissions.

In preparation for model evaluation, the entire data base was examined to establish suitable case-study days. In all, 20 days were selected for simulations based on high observed O_3 maxima and adequate availability of data. The maximum observed O_3 on these days ranged from 160 to 260 ppb.

The three urban models (PBM, LPM, and UAM) were adapted to access the RAPS data archive, which included a detailed emissions inventory with resolution to 1 h and 1 km (Littman, 1979). Although precise input requirements differ among the models, effort was made to supply information in a consistent form for all. Thus, the simulations were done in parallel, using the same basic data sets, and the results give a comparison among models as well as an indication of accuracy against observed concentrations.

As examples of model results, Figures 6 and 7 show observed and predicted O_3 values for the PBM and LPM on a single day, 1 October, 1976. Stagnation conditions prevailed on that day. For the PBM, the time series show concentrations within a fixed box 20 km x 20 km over the city center. Maximum and minimum observed

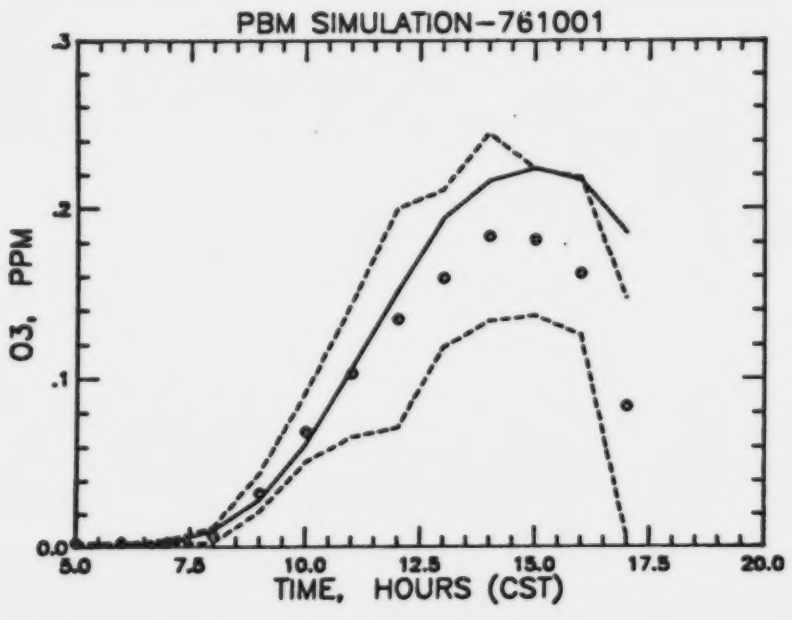

Fig. 6. PBM predicted ozone (solid) compared to measured domain average (circles) and maximum and minimum within the domain (dashed). This result is for the 1 October 1976 RAPS simulation.

concentrations over all the stations (13) in the box are given by the dashed lines. The average observed concentration is given by circles, and the model prediction is given by the solid line. For the LPM, the concentrations refer to a moving parcel with initial horizontal dimensions 5 km x 5 km. Predicted values are given for two vertical levels in the parcel, the surface (L-1) and about 200 m (L-3). This particular parcel arrives at the station (102) observing the maximum 1-h O_3 value at the time of that observation (1400 CST).

The ability of the models to reproduce observed O_3 maxima over the 20 test days is summarized by statistics given in Table 1. Complete results of the urban model evaluation are now available in report form (Schere and Shreffler, 1982).

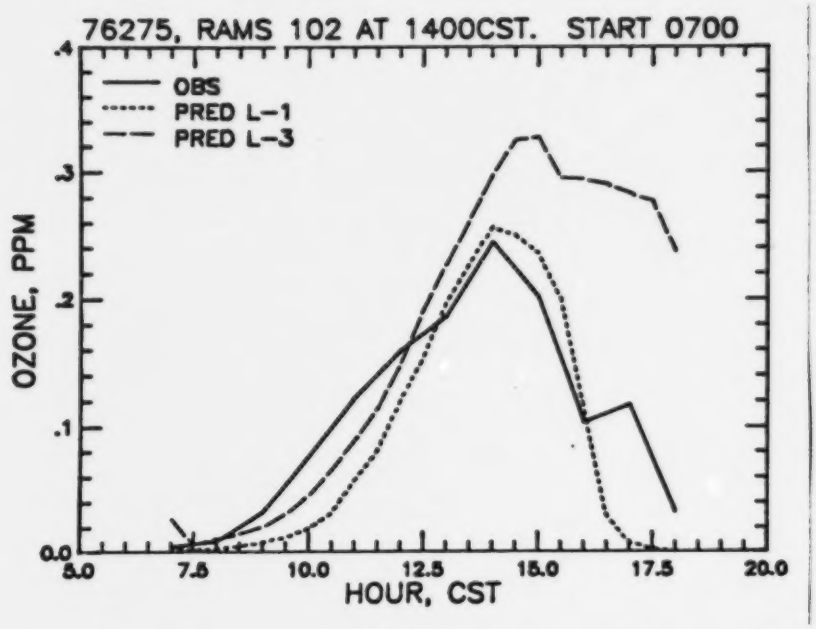

Fig. 7. LPM predicted ozone at the surface (L-1) and 200 m (L-3) compared to the observed ozone. This result is for the 1 October 1976 RAPS simulation.

Table 1. Statistics on residual concentrations (observed minus predicted) of maximum ozone (ppb) from 20 days of RAPS data. The observed maxima ranged 160-260 ppb.

	PBM	LPM	UAM
$\overline{\Delta C}$	-12	5	62
s.d.(ΔC)	39	58	35
$\overline{\lvert\Delta C\rvert}$	29	50	62

A REGIONAL SCALE (1000 KM) MODEL

The U.S. EPA is presently developing a model that can guide the formulation of regional emissions control strategies by estimating the effect of sources on concentrations in remote regions, determining the pollution burden that cities impose on distant neighbors, and eventually analyzing the effect of emissions on acid rain, visibility and fine particles. The utility and credibility of the model will be determined primarily by the extent to which it accounts for all the governing physical and chemical processes. Accordingly, the model is formulated, in principle, to treat all of the chemical and physical processes that are known, or presently thought, to affect the concentrations of air pollutants over several day/1000 kilometer scale domains. Among these processes are (not necessarily in order of importance):

1. Horizontal transport;
2. Photochemistry, including the very slow reactions;
3. Nighttime chemistry of the products and precursors of photochemical reactions;
4. Nighttime wind shear, stability stratification, and turbulence "episodes" associated with the nocturnal jet;
5. Cumulus cloud effects - venting pollutants from the mixed layer, perturbing photochemical reaction rates in their shadows, providing sites for liquid phase reactions, influencing changes in the mixed layer depth, perturbing horizontal flow;
6. Mesoscale vertical motion induced by terrain and horizontal divergence of the large scale flow;
7. Mesoscale eddy effects on urban plume trajectories and growth rates;
8. Terrain effects on horizontal flows, removal, diffusion;
9. Subgrid scale chemistry processes resulting from emissions from sources smaller than the model's grid can resolve;
10. Natural sources of HC, NO_x and stratospheric O_3;
11. Wet and dry removal processes, e.g., washout and deposition.

Of the eleven processes listed above, only the first and last have been treated in any detail in the regional scale models of air pollution developed to date. In fact, a review of these models (see, for example, reviews by Drake and Bass in Henderson et al.

1980) reveals that virtually all of the Eulerian type models are in essence simply expanded urban scale models. They account for the physical processes that are active during daylight hours and within 10 km or so of a source, but they neglect both the processes that are important beyond this distance and those that are active at night.

The U.S. EPA has taken the approach of developing a truly regional model, allowing the processes described above to influence the structure of the model, rather than trying to force the processes into an existing urban structure. The original goal of this work was to develop a specific model of regional scale photochemical air pollution. However, as the work progressed and new developments and ideas continually emerged, the need was seen for a general modeling framework within which the various physical and chemical processes that play important roles could be treated in modular form. This would permit ongoing incorporation into the model of state-of-the-art techniques without the need to overhaul the model each time. The structure and modular form of the Regional Oxidant Modeling System (ROMS) are unique for studying regional scale pollution. Complete documentation of the ROMS is being prepared (Lamb, 1982a, 1982b).

When this model development work was initiated some four years ago, an attempt was made to derive from the observational evidence available at that time an estimate of the minimum vertical and horizontal resolutions necessary to describe regional scale air pollution phenomena. The aim was to arrive at the best compromise between the restrictions imposed upon the model by computer time and memory limitations and the need to describe as accurately as possible all of the governing processes cited above. Careful review was made of the NO, O_3 and meteorological data reported in Siple (1977) by the participants of the 1975 Northeast Oxidant Transport Study. The characteristics of the O_3 distribution described in those reports would require at the very least a three-level model--one level assigned to the surface layer, another level to the remainder of the daytime mixed layer, and an additional layer atop the mixed layer. The top level would be used in conjunction with the mixed layer to account for downward fluxes of stratospheric O_3 as well as upward fluxes of O_3 and its precursors into the subsidence inversion layer above. Material that entered this top layer could be transported by winds aloft to areas outside the modeling region; it could enter precipitating clouds and be rained out of the atmosphere; or it could undergo chemical transformation. Representing the subsidence inversion, where cumulus clouds often form under stagnant high pressure conditions, the top level of the model would be instrumental in simulating the chemical sink effect of heterogeneous (within cloud droplets) reactions among O_3, its precursors, and other natural and pollutant species. Including

cloud effects in the model would be especially important in future simulation of SO_2 and sulfates.

Having three layers in a model is insufficient in itself to simulate all relevant phenomena. For example, three layers of constant thickness are incompatible with the spatial and temporal variability that the radiation inversion and mixed layer thickness are known to have. What is needed in the model is three "dynamic" layers that are free to expand and contract locally in response to changes in the phenomena they are intended to treat. The model discussed here possesses this property. Figure 8 illustrates the vertical structure of this model and the physical phenomena that each layer is intended to simulate. The surfaces that comprise the interfaces of adjacent layers in our model are variable in both space and time in order that each layer can keep track of the changes that occur in the particular set of phenomena that layer is designed to describe (summarized in Figure 8). A consequence of this structure is that the volumes of the grid cells vary in both space and time. By contrast, in conventional models the grid network and cell volumes remain fixed and surfaces such as the mixed layer top move through the grid system. In the following several paragraphs, we elaborate on some of the phenomena cited in Figure 8 that our model will take into account.

During the day the highest layer shown in Figure 8a represents the synoptic scale subsidence inversion, which may or may not contain cumulus clouds. Stratospheric O_3 is transported downward through this layer and anthropogenic O_3 and its precursors can be carried into it by cumulus clouds or penetrative convection. The base of this layer is normally 1 to 2 km above ground level. Below it pollutants are kept well mixed vertically by turbulent convection. If the winds are strong or the surface heat flux is weak, wind speed and direction may vary appreciably within the first several hundred meters above ground. There is usually also a marked difference in the wind speed and direction between the inversion layer and the mixed layer below. Over large lakes and along sea coasts there is frequently a second inversion layer below that generated by synoptic scale subsidence. This lower inversion is produced by sea or lake breeze regimes, and it restricts the vertical mixing of pollutants emitted over the water and within several kilometers inland from the water's edge.

Air drawn into young cumulus clouds originates primarily in the lower portion of the mixed layer. Fresh emissions of NO_x and HC can be transported by the vertical currents that feed these clouds from ground-level to altitudes well above the top of the mixed layer in one steady, upward motion. In the process little or no mixing with aged pollutants in the mixed layer occurs. At night, cumulus clouds usually evaporate, and when they do they

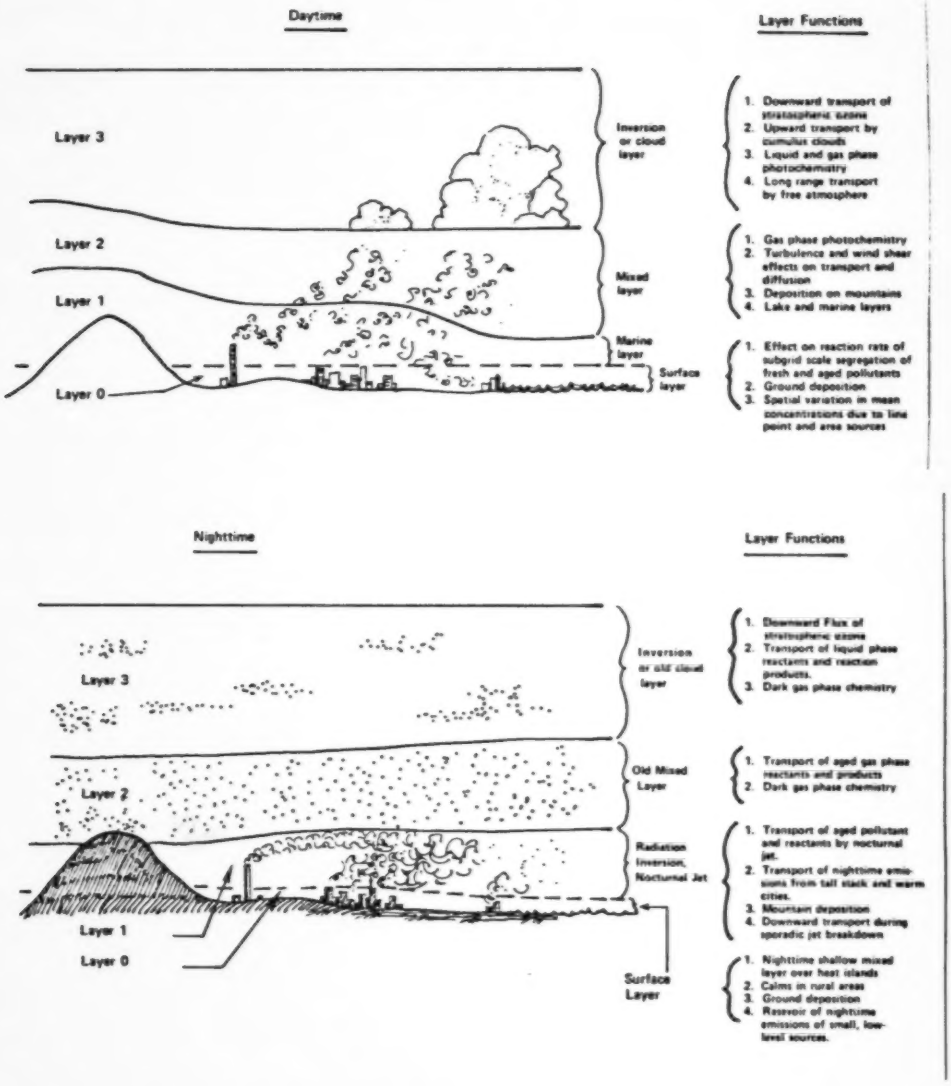
Daytime
Layer Functions
Layer 3
Layer 2
Layer 1
Layer 0
Inversion or cloud layer
Mixed layer
Marine layer
Surface layer
1. Downward transport of stratospheric ozone
2. Upward transport by cumulus clouds
3. Liquid and gas phase photochemistry
4. Long range transport by free atmosphere
1. Gas phase photochemistry
2. Turbulence and wind shear effects on transport and diffusion
3. Deposition on mountains
4. Lake and marine layers
1. Effect on reaction rate of subgrid scale segregation of fresh and aged pollutants
2. Ground deposition
3. Spatial variation in mean concentrations due to line point and area sources
Nighttime
Layer Functions
Layer 3
Layer 2
Layer 1
Layer 0
Inversion or old cloud layer
Old Mixed Layer
Radiation Inversion, Nocturnal Jet
Surface Layer
1. Downward Flux of stratospheric ozone
2. Transport of liquid phase reactants and reaction products.
3. Dark gas phase chemistry
1. Transport of aged gas phase reactants and products
2. Dark gas phase chemistry
1. Transport of aged pollutant and reactants by nocturnal jet.
2. Transport of nighttime emissions from tall stack and warm cities.
3. Mountain deposition
4. Downward transport during sporadic jet breakdown
1. Nighttime shallow mixed layer over heat islands
2. Calms in rural areas
3. Ground deposition
4. Reservoir of nighttime emissions of small, low-level sources.

Fig. 8. Schematic illustration of the dynamic layer structure of the regional model and phenomena each layer is designed to treat: (a) daytime, (b) nighttime.

leave behind products of liquid phase reactions that can be transported hundreds of kilometers before sunrise.

Dramatic changes occur in the mixed layer at night. With the onset of surface cooling following sunset, a stable layer of air forms near the ground that quenches the vertical momentum fluxes that give rise to frictional drag on the horizontal flow. With retardation forces eliminated, the wind just above the stable layer accelerates giving rise to the phenomenon known as the nocturnal jet. Wind speeds in the core of the jet, which usually lies between 300 and 500 meters above ground, may be 10-15 m/s while at the same time the air is nearly calm at the surface. Emissions from tall stacks and from sources within the urban heat island enter the jet region at night. There they react with aged pollutants from the previous day and are transported considerable distances by the strong flow. The remnant of the previous day's mixed layer above the jet is isolated from the influence of fresh emissions and it moves at a slower speed than air below.

Sporadic episodes of turbulence in the shear layer beneath the nocturnal jet are a mechanism by which O_3 and constituents of urban plumes are brought to ground-level at night. There, deposition on surfaces and reactions with emissions of small, low-level sources occur. This sporadic mixing process is perhaps the only mechanism by which the reservoir of aged pollutants aloft can be depleted at night.

One point that we wish to emphasize here is that one-layer regional scale air pollution models are incapable of simulating the effects on pollutants like O_3 of the vertical segregation of aged and fresh emissions that occurs at night. Being cut-off from contact with the ground and fresh NO_x emissions, O_3 above the nighttime radiation inversion is free to travel great distances before it is mixed vertically by convection the following day. The effect of this nighttime segregation of pollutants is to extend greatly the effective residence times of species like O_3 in the lower troposphere. Consequently, a multi-layered model seems to be essential to simulate accurately the long range transport of photochemical air pollutants.

As now planned, the horizontal resolution of the model is about 18 km. The resolution should be as high as possible to mitigate the effects of subgrid scale concentration fluctuations. A scheme to treat subgrid chemistry is implemented in Layer 0, adjacent to the ground. Layer 0 is treated diagnostically in the governing equations, and also handles surface depletion. In modeling atmospheric processes over 1000 km scale regions, effect of the earth's curvature must be taken into account. Model equations are transformed into a curvilinear frame in which latitude, longitude and

elevation are coordinates and the basis vectors point north, east and vertically upward at every point on the earth. We have chosen this frame because it is a natural one from which transformations to any rectilinear system are easily performed. Also, is the frame in which worldwide meteorological data are reported.

There are three basic problems that must be overcome to make operational a model as large and comprehensive as the one we are developing.

First, due to the large number of processes that we plan to treat, our model is rather complicated. In order to alleviate the problems that this might cause in operating the model and in making future refinements, we have structured it so that its central core consists solely of a set of algorithms for solving the coupled set of generalized finite difference equations that describe processes in each of its layers. The modeling functions of describing the mixed layer dynamics; topographic effects on winds; chemistry; cloud fluxes, etc. will be handled by a set of special processors that are external to the central model and which feed the model key variables through a computer file. Within this framework the techniques used to describe the various physical processes can be altered without overhauling the model itself. An additional advantage is that execution times are greatly reduced when several runs of a given scenario are to be performed in which only one or a few parameter values are altered.

A second problem is limitations of computer storage capacity. To simulate air quality over the northeastern United States with the horizontal resolution we desire, our model has roughly 10^4 grid points and treats 25 (eventually more) chemical species. Thus, the concentration variables alone require 250K words of storage and this is just under the working limit of 260K words of memory on EPA's Univac computer. To accomodate a model of the anticipated size we have developed special techniques for handling the modeling domain in piecewise fashion.

Finally, the empirical data needed to parameterize some of the physical phenomena cited earlier are not presently available. To remedy this, EPA initiated project NEROS (Northeast Regional Oxidant Study) to collect during the summers of 1979 and 1980 the meteorological and chemical data required to formulate the model. A second goal of NEROS was to gather the data required to perform comprehensive test runs and evaluation exercises of the model (see Clark and Clarke, 1982; Clarke et al. 1982).

Aircraft sampling during NEROS was designed to gather evaluation data for the regional model. Using a trajectory model and radiosonde data, flight tracks were directed to obtain Lagrangian

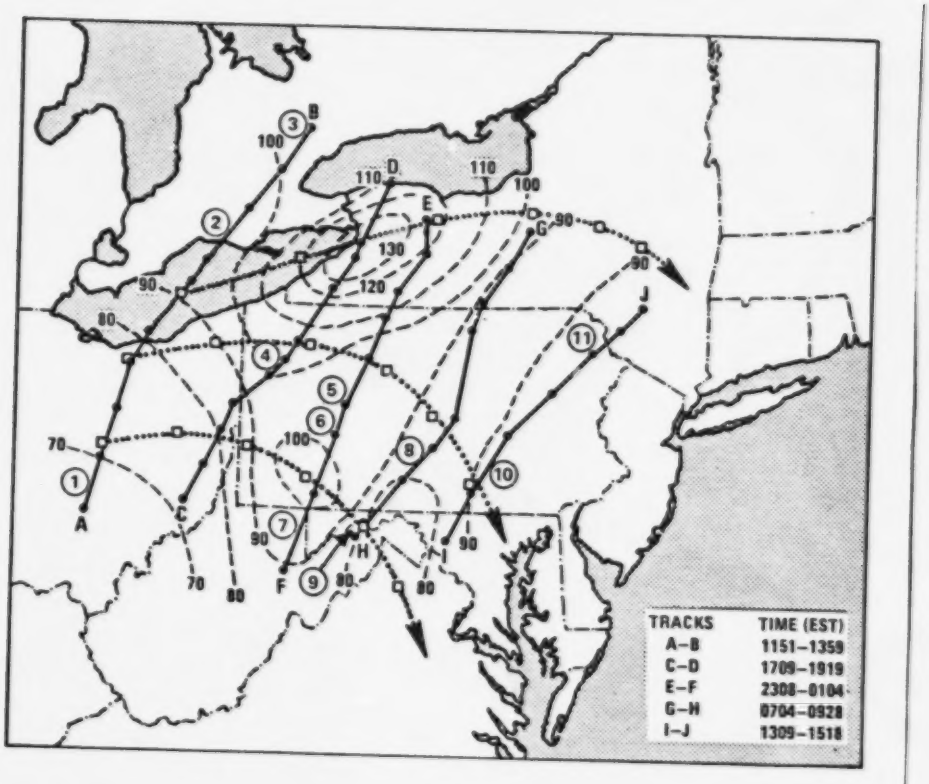

Fig. 9. Aircraft flight tracks, ozone concentrations (ppb), and air trajectories (6-hr segment given by (□)) for 3-4 August 1979.

sampling of an air mass. Figure 9 shows computed trajectories and corresponding aircraft flight tracks with O_3 concentration isopleths on one day. Figure 10 gives the O_3 concentrations in a vertical cross-section along one of the flight tracks (E-F). The significant O_3 concentrations at the level of the nocturnal jet are evident in these data taken near midnight.

The model is currently set up with a 60 x 42 array of grid cells. Figure 11 shows the region of the northeast U.S. being modeled as well as NO_2 isopleths resulting from a short test run. Although the model is functioning, substantial development work must yet be accomplished. Preprocessors must be thoroughly checked for accuracy, a refined emissions inventory (including biogenics) must be finished, and the NEROS data base must be analyzed. We

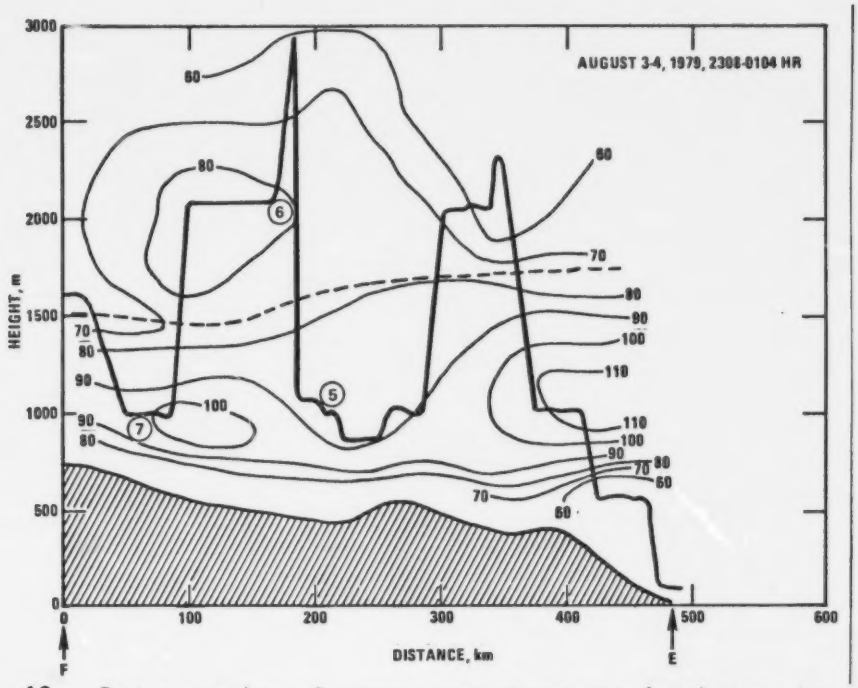

Fig. 10. Cross-section of ozone concentrations (ppb) for the track E-F in Fig. 9. Aircraft path is given by solid line, and the dashed line shows an elevated inversion.

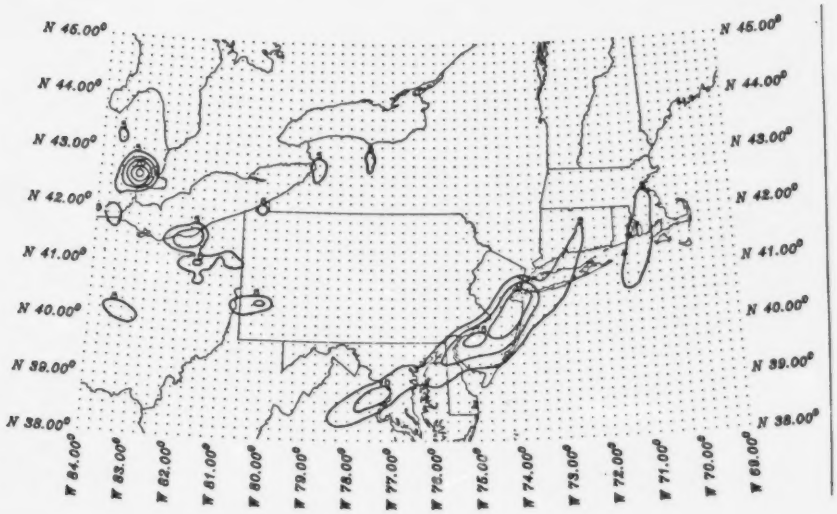

Fig. 11. The grid of the regional model and NO_2 isopleths from a test run. The isopleths are for 0800 local time and reflect sources about urban areas.

believe these tasks can be completed in mid-1983. After that we would need about another year of model running and evaluation before the model would be operational and could conceivably be transferred to other user groups (perhaps mid-1984).

CONCLUDING REMARKS

Over the past decade, focus of research, development, and evaluation efforts concerning photochemical models has been initially on the urban O_3 problem and more recently on the regional O_3 problem. Because of the earlier emphasis on the urban scale, there now exist in operational form several PAQSM's which are designed to simulate urban photochemistry and which have been applied in the U.S., Europe, and Australia. We have summarized the attributes of three such models which have emerged from programs of the U.S. EPA and been evaluated with a comprehensive urban data base. The models have been shown to be effective in predicting urban O_3 concentrations. The choice of which model to use will depend, in part, on the spatial resolution required and the computer resources available.

The impetus for development of a regional model resulted from a growing awareness that constituents of photochemical smog travel long distances, and control of locally generated O_3 may not be sufficient to alleviate air quality problems. On the regional scale, the multi-day nature of long-range transport necessitates consideration of factors which may be neglected for a one-day episode on the urban scale. These processes include slow photochemical reactions and segregation of surface and upper layers at night. Testing and evaluation of the regional model using data of the NEROS program will begin in 1983.

In confining our review to progress with EPA models, we do not mean to exclude other approaches which are being considered but believe that whatever choices are made the number of basic frameworks must remain quite limited due to the large resource requirements of PAQSM's. Thus, refinement of a single approach is preferred over a proliferation of models with slightly different characteristics. We have emphasized that successful application of a PAQSM involves the use of an extensive, and usually expensive, data base including information on meteorology, air quality, and source emissions. We believe that commitment to gathering such data should stand equally with efforts at developing the mathematical constructs and computer programs which constitute the PAQSM.

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